Sourcing obsidian artefacts from Early Neolithic sites in south-central Romania

SOURCING OBSIDIAN ARTEFACTS FROM EARLY NEOLITHIC SITES IN SOUTH-CENTRAL ROMANIA

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Keywords: *pXRF, obsidian sourcing, Neolithic, Starčevo-Criș, south-central Romania*
Abstract: *Portable X-ray fluorescence spectroscopy (pXRF) was used to reveal the chemical signatures of 60 obsidian artefacts from two Early Neolithic sites in the Muntenia region of southern Romania – Uliești in Dâmbovița County and Măgura – Buduiasca in Teleorman County. The results show that the Starčevo-Criș communities at both sites used obsidian that originated from geological sources in the Carpathians. Obsidian from the C1 and C2 source areas occurs at Măgura, while only C1 obsidian has been documented in the much smaller assemblage from Uliești. We consider the implications of these results for obsidian procurement patterns documented among the earliest farmers of the northern Balkans.*

Cuvinte-cheie: *pXRF, surse de obsidian, neolitic, Starčevo-Criș, sud-centrul României*
Rezumat: *Spectrometrul portabil cu raze X (pXRF) a fost utilizat pentru a determina semnătura chimică a 60 de piese din obsidian provenind din două situri neolitice timpurii din Muntenia, sudul României: Uliești din județul Dâmbovița și Măgura – Buduiasca din județul Teleorman. Conform rezultatelor obținute, comunitățile Starčevo-Criș din ambele localități au folosit obsidian care provenea din surse geologice carpatice. Obsidianul din zonele sursă C1 și C2 apare la Măgura, în timp ce numai obsidianul de tip C1 a fost documentat în ansamblul litic mult mai redus de la Uliești. Sunt discutate apoi implicațiile acestor rezultate pentru modelele de obținere a obsidianului de către comunitățile neolitice timpurii din zona nord-balcanică.*

INTRODUCTION

Geochemical fingerprinting is acknowledged to be the most accurate means of determining the provenance of lithic raw materials used by prehistoric societies and has been used extensively in obsidian research in Europe and the Near East. In this paper we present the results of a geochemical characterization study of obsidian artefacts from two Early Neolithic sites (Uliești and Măgura – *Buduiasca*) in the Muntenia region of southern Romania.

While over fifty Early Neolithic sites have been recorded in south-central Romania (Muntenia and Oltenia east of the Jiu River valley), less than one-third have been excavated systematically and obsidian has been reported from only nine sites, invariably in only very small quantities. This situation is mirrored on the Danube Plain of northern Bulgaria where obsidian has been reported from just two Early Neolithic sites, again in very small amounts (Table 1; Fig. 1). The scarcity of obsidian in Early Neolithic contexts in the Lower Danube Basin contrasts with the prominence of "Balkan flint" in Early Neolithic chipped stone assemblages across the region. Distance to source and "competition" from other high-quality lithic resources were perhaps factors influencing this pattern. Obsidian sources in the Carpathian Mountains in Hungary and Slovakia are 500–700 km distant from the sites considered here, while the Balkan flint sources along the Danube at Nikopol in Bulgaria and Ciuperceni in Romania are very much nearer.

SITES AND SAMPLES

Uliești (Dâmbovița County)

Surface archaeological survey in Uliești commune, ca. 37 km south of Târgoviste, identified traces of Early Neolithic settlement on an elevated alluvial terrace above the River Neajlov. The richest concentration of finds occurred in an area measuring approximately 150 × 70 m centred on 44°34'37.21" N, 25°25'38.99" E, ca. 650 m east of the village of Croitori; the finds included lithic artefacts and sherds of chaff-tempered pottery typical of the Starčevo-Criș culture. A second artefact concentration was found ca. 700 m downstream on the same terrace feature (44°34'16.16" N, 25°26'6.22" E), opposite the village of Corbii Mari – Petrești, and comprised a few Starčevo-Criș-type sherds and lithics, mixed with material of medieval to modern date. Among the lithic artefacts recovered from the two artefact scatters were 22 made of imported "Balkan flint" and four of obsidian – three from Uliești – Croitori and one from Corbii Mari – Petrești (Fig. 2). From the characteristics of the pottery sherds, the Early Neolithic finds from Uliești and Corbii Mari were attributed to the Starčevo-Criș III phase (Ilie, Niță 2014, p. 64).

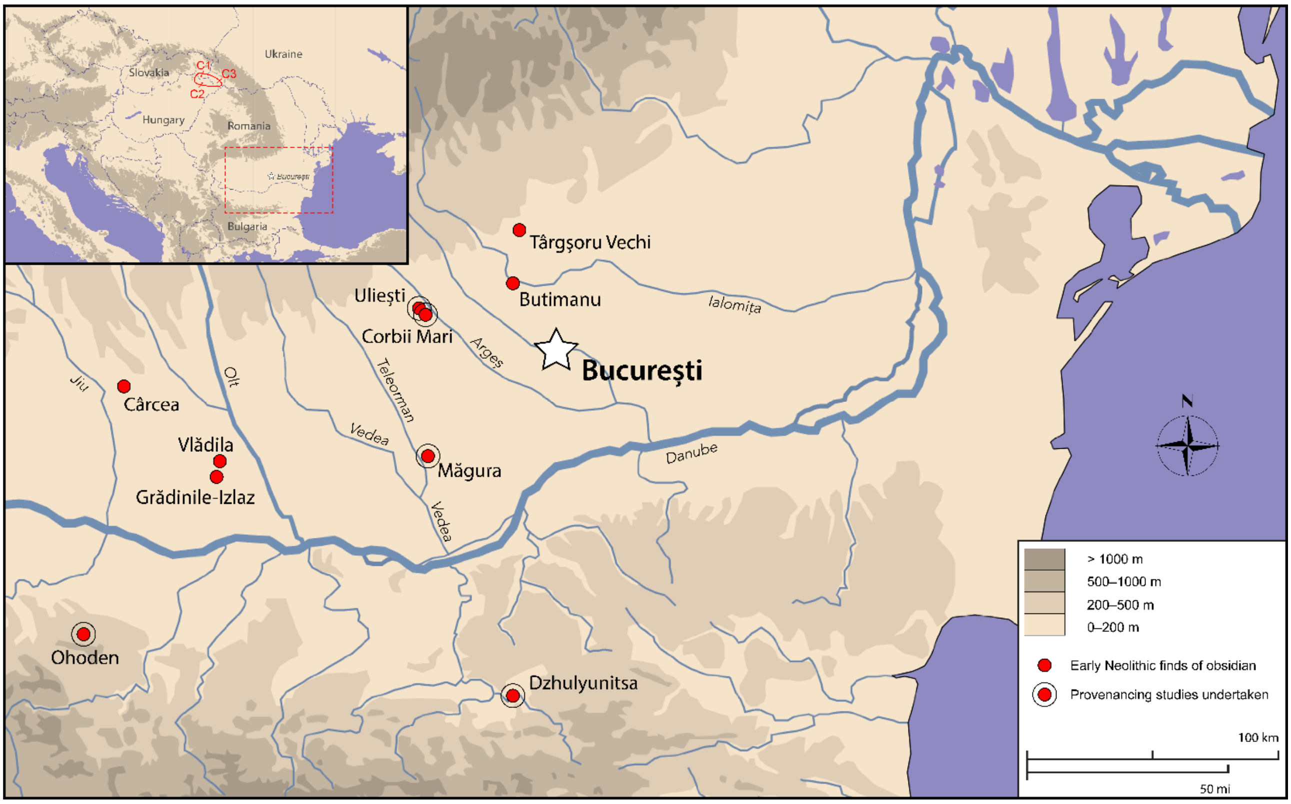

Figure 1. *Early Neolithic sites with obsidian in the Lower Danube Basin, east of the Jiu River valley. The inset map shows the location of the study region in relation to obsidian source areas in the Western Carpathians.*

Site	**County**	**Province**	**Geographical coordinates**	**Obsidian as a proportion of the lithic assemblage**	**References**
Târgşoru Vechi	Prahova	Muntenia	44°52'37.01" N, 25°55'09.71" E	?	Păunescu 1970
Ulieşti	Dâmboviţa	Muntenia	44°34'37.21" N, 25°25'38.99" E	?	Ilie, Niţă 2014
Corbii Mari	Dâmboviţa	Muntenia	44°34'16.16" N, 25°26'6.22" E	?	Ilie, Niţă 2014
Butimanu	Dâmboviţa	Muntenia	44°40'15.60" N, 25°52'48.62" E	?	Comşa 1969
Măgura – *Buduiasca*	Teleorman	Muntenia	44°01'02.14" N, 25°24'41.26" E	~1%	Andreescu, Mirea 2008
Cârcea – *La Hanuri*	Dolj	Oltenia	44°17'11.19" N, 23°53'02.85" E	<2%	Dinan, Nica 1995; Păunescu 1988; Nica 1976
Cârcea – *Viaduct*	Dolj	Oltenia	44°16'07.30" N, 25°53'07.44" E	4%	Dinan, Nica 1995
Vlădila	Olt	Oltenia	43°59'56.38" N, 25°24'11.18" E	<1%	Dinan, Nica 1995
Grădinile – *Izlaz*	Olt	Oltenia	43°56'45.77" N, 25°23'32.25" E	<2%	Dinan, Nica 1995; Păunescu 1988, Nica 1981

Table 1. *Early Neolithic occurrences of obsidian in south-central Romania (acc. to Fig. 1).*

Figure 2. *Obsidian artefacts from Uliești – Croitori (1–3) and Corbii Mari – Petrești (4). Attributes of the piece from Corbii Mari (not included in Table 2) are Type = retouched flake, Length = 28 mm, Breadth = 19 mm, Thickness = 6 mm, Weight = 3.18 g.*

Figure 4. *Obsidian artefacts from Măgura: nos. 1–3 (TL.06, TL.08, TL.09 – C2E obsidian, from Bodul lui Moș Ivănuș); no. 4-5 (TL.24, TL.23, C1 obsidian, from Buduiasca).*

Măgura – *Buduiasca* (Teleorman County)

The site of Măgura – *Buduiasca* (44°01′02.14″ N, 25°24′41.26″ E), also known as *Teleor 3*, is situated on the eastern edge of the village of Măgura in the Teleorman Valley, ca. 8 km northeast of the town of Alexandria and ca. 45 km above the confluence of the River Vedea with the Danube. The site covers an area of about 30 ha on a Late Pleistocene alluvial terrace about 8 m above the river level. Around 400 m^2 of the site were excavated between 2001 and 2008 (Andreescu, Mirea 2008; Mirea 2011).

The earliest Neolithic (Starčevo-Criș I) occupation at Măgura – *Buduiasca* occurred on a slight elevation known as *Boldul lui Moș Ivănuș* – possibly a remnant of an older terrace feature. During a later phase of the Early Neolithic (Starčevo-Criș III) the settlement expanded across the entire Buduiasca site area and was succeeded by Middle Neolithic (Dudești culture) and Late Neolithic (Vădastra culture) occupations. Evidence from stratigraphy, typology and single-entity ^{14}C dating suggests the following Neolithic occupation sequence and chronology for Măgura – *Buduiasca*: Starčevo-Criș I – ca. 6000–5800 cal BC, Starčevo-Criș III – ca. 5750–5600 cal BC, Dudești – ca. 5600–5300 cal BC and Vădastra – ca. 5300–5175 cal BC (Fig. 4).

A total of 59 obsidian artefacts were recovered in the 2001–2008 excavations at Măgura – *Buduiasca* (Fig. 3), 57 of which are considered in this paper (two tiny obsidian "chips" were considered too small to yield reliable results using pXRF). Of these 57 specimens, all but four (n = 53) were excavated from *Boldul lui Moș Ivănuș* – 23 were recovered from well-defined archaeological features belonging to the Criș I phase (variously interpreted as pits, house or hut foundations re-used as pits, or agglomerations of archaeological material), ten came from the Criș I "cultural layer", and the remaining 20 pieces came from "mixed" or disturbed contexts. Of the four obsidian artefacts found elsewhere on the Măgura – *Buduiasca* site, two came from well-defined features belonging to the Criș III phase, one came from the Criș III "cultural layer", and one from a "mixed" or disturbed context (Table 2).

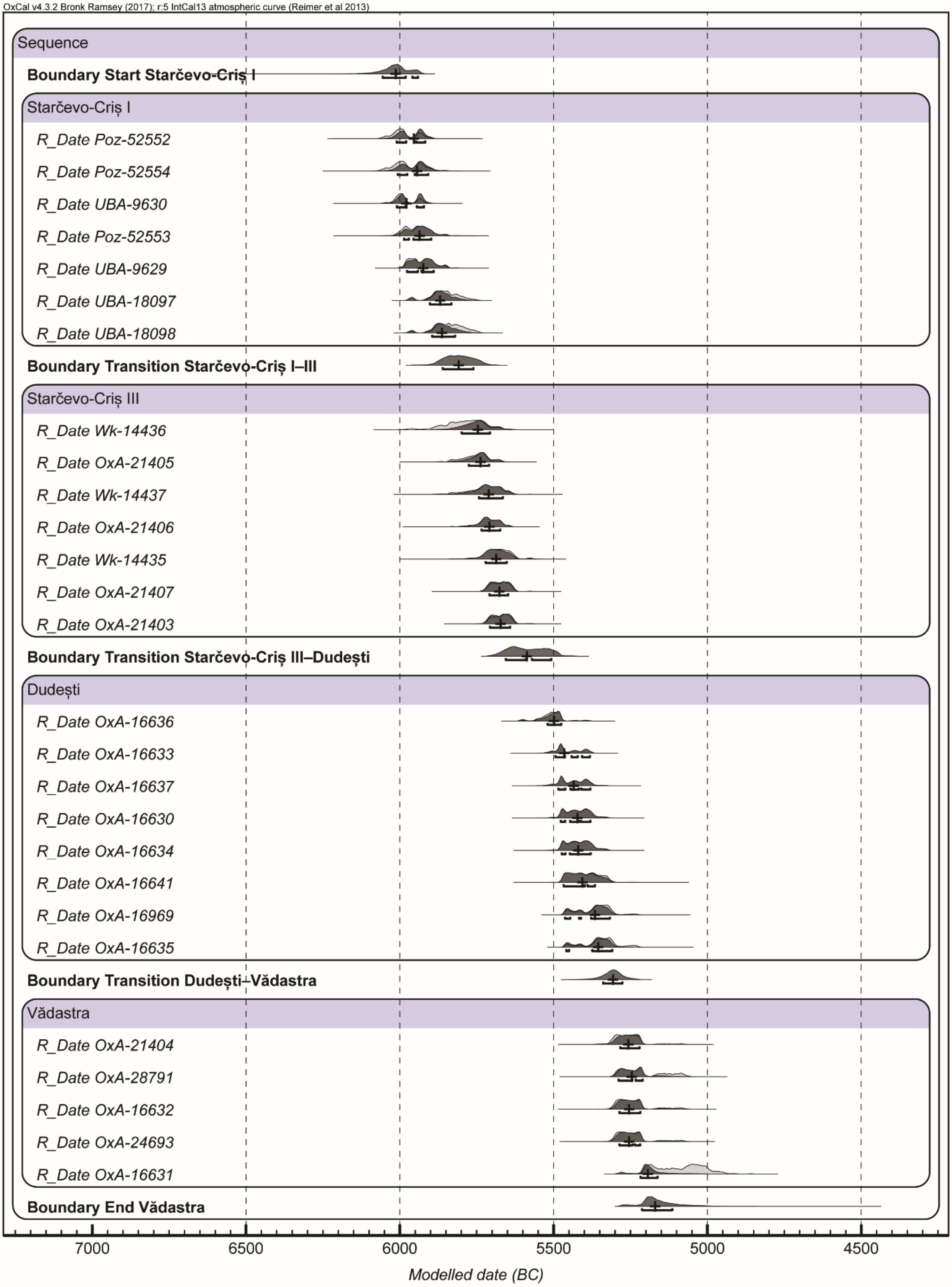

Figure 3. *Bayesian chronological model of Neolithic occupation phases at Măgura – Buduiasca implemented in OxCal v 4.3.2 (Bronk Ramsey 2009) using the IntCal13 dataset (Reimer et alii 2013).* ^{14}*C data from Mărgărit et alii 2018, table 1. Probability distributions of the calibrated dates are shown in light-grey. Dark-grey distributions are posterior density estimates of the dates of samples included in the OxCal models, and of the beginning and end of each phase (OxCal Boundary).*

Find No.	Year	Trench, Grid Sq., Level	Context	Phase	Type	Blank	L	B	Th	Wgt	Cortex	Sample
Uliești-Croitori												
79.074.005, piesa #1			Surface find	Criș III	Unretouched bladelet (distal mesial break)	B	31.3	8.7	2.3	0.7		**UC.01**
79.074.005, piesa #2			Surface find	Criș III	Unretouched bladelet	B	20.3	9.8	2.7	0.5	x	**UC.02**
79.074.005, piesa #3			Surface find	Criș III	Bipolar core/"wedge"?	F?	26.9	16.8	9.7	3.3		**UC.03**
Măgura-Boldul lui Moș Ivănuș												
UN 2944	2008	S51, □B1, -1.00-1.10	Criș I layer	Criș I	Unretouched flake	F	n.r.	n.r.	n.r.	1.08		**TL.01**
UN 2936	2008	S51, □A1, -0.90-1.00	Criș I layer	Criș I	Irregular flake/Core fragment	F	n.r.	n.r.	n.r.	1.99		**TL.02**
UN 2800	2007	S47, □A1.2, -1.20-1.30	Cpl. 56	Criș I	Irregular flake/Core fragment	F	n.r.	n.r.	n.r.	4.49		**TL.03**
UN 2854	2007	S48, □B1, -1.30-1.50	Mixed layer, overlying Cpl. 55 & 57 (50% Criș and 50% Dudești)	Criș I?	Irregular flake/Core fragment	F	n.r.	n.r.	n.r.	4.52	x	**TL.04**
UN 3030	2008	S51, □A1.4, -1.30-1.40	Cpl. 58	Criș I	Unretouched flake (distal break)	F	n.r.	n.r.	n.r.	1.77	x	**TL.05**
UN 3055	2008	S51, □B.3.3, -1,30-1,40	Cpl. 51	Criș I	Unretouched blade	B	39.31	19.76	4.65	2.80		**TL.06**
UN 3105	2008	S51, □B.1.2, -1,60-1,70	Cpl. 58	Criș I	Edge-retouched blade	B	38.11	14.90	4.23	1.69		**TL.07**
UN 2955	2008	S52, □D.1, -0,90-1,10	Criș I layer	Criș I	Unretouched blade (mesial fragment)	B	34.39	12.35	2.38	1.30		**TL.08**
UN 3147	2008	S52, □B.3, -1,40-1,50	Criș I layer	Criș I	Unretouched flake	F	27.72	14.59	3.54	1.57		**TL.09**
UN 2664	2006	S37, □C.1.2, -0,70-1,07	Gr. 12 (medieval pit with Criș I intrusions)	Criș I?	Unretouched flake	F	25.79	15.00	6.36	2.10		**TL.10**
UN 2859	2007	S50, □1, -1,40-1,50	Mixed layer, overlying Cpl. 55 & 57 (50% Criș and 50% Dudești)	Criș I?	Unretouched bladelet (distal mesial break)	B	20.66	7.51	1.71	0.36		**TL.11**
UN 2790	2007	S48, □1, -1,20-1,30	Mixed layer, overlying Cpl. 55 & 57 (50% Criș and 50% Dudești)	Criș I?	Unretouched bladelet (distal mesial break)	B	21.89	10.06	2.64	0.40		**TL.12**
UN 2771	2007	S48, □2, -1,00-1,15	Cpl. 50	Criș I	Bladelet (unretouched distal fragment)	B	18.97	8.07	1.83	0.22		**TL.13**
UN 2754	2007	S46, □A1.2, -1,75-1,83	Cpl. 49	Criș I	Unretouched blade	B	23.53	10.78	2.12	0.47		**TL.14**
UN 2681	2007	S46, □4, -0,90-1,10	Mixed layer (but mainly Criș I)	Criș I?	Unretouched flake	F	14.24	12.54	2.93	0.90		**TL.15**
UN 2957	2008	S52, □C.1, -0,90-1,00	Mixed layer (but mainly Criș I)	Criș I?	Unretouched bladelet (distal mesial break)	B	18.80	11.20	2.61	0.76		**TL.16**

Find No.	Year	Trench, Grid Sq., Level	Context	Phase	Type	Blank	L	B	Th	Wgt	Cortex	Sample
UN 2845	2007	S50, □1, -1,30-1,40	Mixed layer, overlying Cpl. 55 & 57 (50% Criș and 50% Dudești)	Criș I?	Unretouched bladelet	B	25.39	9.54	3.18	0.72		**TL.17**
UN 3059	2008	S52, □A.1, -1,20-1,30	Cpl. 60	Criș I	Unretouched flake	F	23.60	12.89	3.31	0.95		**TL.18**
UN 2876	2007	S48, □A1.2-B.1.2, -1,80-1,85	Cpl. 57	Criș I	Unretouched flake	F	21.85	12.30	3.06	0.63		**TL.19**
UN 3082	2008	S51, □A.1.1, -1,60-1,70	Cpl. 58	Criș I	Unretouched flake	F	20.73	13.20	3.35	0.67		**TL.20**
UN 2464	2006	S34, -0,70-0,80	Criș I layer	Criș I	Unretouched flake	F	16.62	12.15	3.43	0.61		**TL.21**
UN 2722	2007	S47, □1, -0,90-1,00	Cpl. 50	Criș I	Unretouched blade (mesial fragment)	B	19.89	12.72	2.85	0.94		**TL.22**
UN 1340	2004	S22, □B2, -1,00-1,10	Criș III layer, overlying Cpl. 35 (Criș III)	Criș III	Unretouched bladelet	B	26.06	8.76	2.47	0.59		**TL.23**
UN 488	2003	S10A, □A1, -0,90-1,00	At the top of Cpl. 13 (Criș III)	Criș III	Unretouched flake	F	20.34	17.74	2.54	0.75		**TL.24**
UN 573	2003	S10D, □D3, -1,50-1,60	At the base of Cpl. 15/16 (Dudești) overlapping a thin Criș III layer	Criș III?	Unretouched flake	F	20.88	16.39	4.83	1.38		**TL.25**
UN 1552	2004	S20, □B.1.6, B.2.4, -1,20-1,30	Mixed layer Criș III, Dudești, Vădastra, overlapping a Vădastra complex (Cpl. 22)	Criș III?	Unretouched bladelet	B	34.06	11.56	8.59	2.45		**TL.26**
UN 2563	2006	S.43 -2,00-2,10	Criș I layer	Criș I	Unretouched flake	F	21.36	25.19	5.20	1.68		**TL.27**
UN 2860	2007	S50, □2, -1,40-1,50	Criș I layer, overlying Cpl. 49 & 58	Criș I	Unretouched flake	F	11.21	15.56	4.33	0.78		**TL.28**
UN 3194	2008	S52, □B.2.2, -1,80-1,90	Cpl. 57	Criș I	Unretouched flake	F	17.17	17.60	3.60	0.78	x	**TL.29**
UN 2859	2007	S50, □1, -1,40-1,50	Mixed layer, overlying Cpl. 55 & 57 (50% Criș and 50% Dudești)	Criș I?	Unretouched bladelet	B	22.49	8.91	3.76	0.82	x	**TL.30**
UN 3151	2008	S52, □B.2.1-B2.3, -1,50-1,60	Mixed (50% Criș and 50% Dudești)	Criș I?	Unretouched flake	F	15.43	15.92	7.00	1.55	x	**TL.31**
UN 3191	2008	S52, □B.3.3-C.3.1, -1,80-1,90	Cpl. 57	Criș I	Unretouched flake	F	16.95	12.55	2.32	0.35		**TL.32**
UN 2713	2007	S47, □3, -0,70-0,95	Mixed layer (Criș & Dudești)	Criș I?	Unretouched flake	F	13.55	14.71	3.99	0.53		**TL.33**
UN 2867	2007	S48, □A.1.2-B.1.1-B.1.2, -1,70-1,75	Cpl. 57	Criș I	Blade (unretouched proximal fragment)	B	11.14	13.21	3.55	0.45		**TL.34**
UN 3157	2008	S52, □A.2, -1,50-1,60	Mixed, but mainly Criș I	Criș I?	Blade (unretouched distal fragment)	B	15.52	12.62	2.96	0.53		**TL.35**
UN 3068	2008	S51, □A.1.2, -1,40-1,50	Cpl. 58	Criș I	Unretouched flake	F	14.76	16.03	3.76	0.61		**TL.36**
UN 2711	2007	S46, □A.1.2, -1,55-1,60	Cpl. 49	Criș I	Unretouched flake	F	17.69	15.47	6.95	1.41		**TL.37**
UN 3045	2008	S52, □B.1, -1,20-1,30	Cpl. 60	Criș I	"Wedge"/Pièce esquillée?	F	12.26	11.38	4.35	0.55		**TL.38**

Find No.	Year	Trench, Grid Sq., Level	Context	Phase	Type	Blank	L	B	Th	Wgt	Cortex	Sample
UN 2966	2008	S51, □A.1.1, -1,10-1,20	Criș I layer	Criș I	Unretouched flake	F	15.84	9.05	1.65	0.27		**TL.39**
UN 2852	2007	S50, □2, -1,10-1,20	Mixed layer, overlying Cpl. 49 & 58 (mainly Criș I)	Criș I?	Unretouched flake	F	15.13	9.28	1.71	0.24		**TL.40**
n.r.	2008	2007 backfill	Mixed	Criș I?	Bladelet (unretouched proximal fragment)	B	15.41	8.21	2.61	0.35		**TL.41**
UN 2710	2007	S47, □4, -0,50-0,70	Mixed layer, mainly Criș I	Criș I?	Unretouched flake	F	11.15	12.23	3.16	0.48		**TL.42**
UN 3070	2008	S51, □B1.2, -1,40-1,50	Criș I layer	Criș I	"Wedge"/Pièce esquillée?	F?	13.49	7.24	5.73	0.63		**TL.43**
UN 2709	2007	S46, □A.2.1-A.2.2, -1,55-1,60	Cpl. 49	Criș I	"Wedge"/Pièce esquillée?	F	11.57	10.72	2.21	0.31		**TL.44**
UN 2779	2007	S48, □2, -1,15-1,25	Cpl. 50	Criș I	Unretouched bladelet	B	17.30	3.95	2.25	0.16		**TL.45**
UN 2856/1	2007	S50, □2, -1,30-1,40	Mixed layer, overlying Cpl. 49 & 58 (mainly Criș I)	Criș I?	Unretouched flake	F	10.92	6.93	1.99	0.14		**TL.46**
UN 2856/2	2007	S50, □2, -1,30-1,40	Mixed layer, overlying Cpl. 49 & 58 (mainly Criș I)	Criș I?	Unretouched flake	F	12.46	13.46	2.48	0.32		**TL.47**
UN 3008	2008	S52, □B.3, -0,70-0,90	Mixed	Criș I?	Bladelet (unretouched distal fragment)	B	11.98	10.42	4.51	0.44		**TL.48**
UN 2956	2008	S52, □D.3, -0,90-1,00	Mixed	Criș I?	Unretouched flake	F	8.83	9.82	2.93	0.24		**TL.49**
UN 3194	2008	S52, □B.2.2 -1,80-1,90	Cpl. 57	Criș I	Unretouched flake	F	7.69	11.23	2.15	0.17		**TL.50**
UN 2685	2007	S46, □3, -0,90-1,10	Mixed layer, mainly Criș I	Criș I?	Unretouched flake	F	12.28	14.30	3.79	0.69		**TL.51**
UN 3030	2008	S51, □A.1.4, -1,30-1,40	Cpl. 58	Criș I	Bladelet (unretouched proximal fragment)	B	12.65	7.54	2.11	0.18		**TL.52**
UN 3144	2008	S52, □A.3, -1,30-1,40	Mixed layer, mainly Criș I	Criș I?	Unretouched flake	F	12.33	9.32	2.82	0.35		**TL.53**
UN 3031	2008	S51, □B.1.2, -1,30-1,40	Cpl. 58	Criș I	Unretouched flake	F	15.04	8.73	2.34	0.22		**TL.54**
UN 3044	2008	S51, □B.3.3, -1,10-1,20	Criș I layer	Criș I	Bladelet (unretouched distal fragment)	B	15.47	8.33	2.65	0.30		**TL.55**
UN 3102/1	2008	S52, □B.1, -1,40-1,50	Cpl. 60	Criș I	Unretouched flake	F	16.59	8.72	2.89	0.42	x	**TL.56**
UN 3102/2	2008	S52, □B.1, -1,40-1,50	Cpl. 60	Criș I	Obliquely truncated bladelet (proximal mesial break)	B	19.29	7.76	3.25	0.51		**TL.57**

Table 2. *Details of obsidian artefacts from Uliești – Croitori (UC) and Măgura – Buduiasca (TL) analysed by pXRF. Abbreviations: B – blade, F – flake, Cpl. – complex; n.r. – not recorded.*

METHODOLOGY AND RESULTS

Non-destructive ED-XRF analyses of 57 obsidian artefacts from Măgura – *Buduiasca* and the three obsidian artefacts from Uliești – Croitori were carried out using a Thermo Scientific *Niton XL3t Ultra* (portable) analyser. This particular instrument is equipped with an Ag anode X-ray source (capable of a maximum voltage of 50 keV, current of 200 µA and power of 2W) and a 45 mm^2 Silicon Drift Detector (SDD). Analyses are performed using beam filters to improve detection of particular elements. The 'XL3t Ultra' has an analytical range of up to 30 elements from Mg to U, although this varies according to the 'mode' (calibration model) selected – the analyser is supplied with a number of in-built factory 'calibrations' optimised for analysis of specific materials. The calibrations/modes provided with the 'XL3t Ultra' that are most suitable for the analysis of obsidian (and other bulk samples) are the 'Mining' and 'Soil' calibration models.

Routinely, we perform *two* sets of measurements on obsidian samples, one set with the instrument operated in the *Fundamental Parameter* (FP) "Mining" mode and the other set using the *Compton Normalization* "Soil" mode. FP and Compton Normalization represent different mathematical approaches to the quantification of XRF spectra from samples. Experience shows the latter approach can reduce problems with the measurement of "thin" samples and can provide data for heavy elements (*e.g.* Th, U) at low concentrations. The energy ranges and filter configurations of the Niton XL3t XRF analyzer when operated in "Mining" and "Soil" modes, and the elements analysed, are shown in Table 3. A third type of calibration, known as *Empirical Calibration*, can be achieved by obtaining readings on samples of known elemental composition (Certified Reference Materials) then using the correlation between the readings obtained on the CRMs and their known values (using linear regression analysis) to derive a calibration factor for each individual element. In this way, the values generated for archaeological samples by the analyser can be "recalibrated" using the empirically-derived calibration factors. While recalibrating against reference standards (in theory) produces more accurate results, it is unlikely to have much impact on one's interpretation of the data.

Table 4 presents the results of analyses of the archaeological obsidians from Uliești – Croitori and Măgura – *Buduiasca* performed using the "Mining" and "Soil" calibration models. Results are presented for 10 elements (Ti, Fe, Zn, Rb, Nb, Sr, Y, Zr, Th, U) that have been found to be particularly useful for obsidian provenancing. The measurement window of the analyser was set to the 8 mm spot size. Each sample was analysed for a total of 180 seconds – 60s using each of the 'Main', 'High' and 'Low' range filters that optimize the analyser's sensitivity for various elements. To improve accuracy, the measurements obtained with the factory-set 'Mining' and 'Soil' calibrations were *recalibrated* against data for 23 CRMs – pressed powder samples of various rock types – which were obtained using identical instrument settings.

Calibration	Energy ranges / Filters			
	Main	Low	High	Light
Mining	Al@50kV	Cu@20kV	Mo@50kV	No Filter@8kV
	Sb, Sn, Cd, Pd, Ag, Mo, Nb, Zr, Sr, U, Rb, Th, Bi, As, Se, Au, Pb, W, Ga, Zn, Cu, Re, Ta, Hf, Ni, Co, Fe, Mn, Cr, V, Ti	Cr V Ti Ca K	Ba, Sb, Sn, Ca, Pd, Ag, Y	Mg, Al, Si, P, S, Cl
Soil	Al@50kV	Cu@20kV	Mo@50 kV	N/A
	Mo Zr Sr U Rb Th Pb Se As Hg Au Zn W Cu Ni Co Fe Mn	Cr V Ti Sc Ca K S	Ba Cs Te Sb Sn Cd Ag Pd	**N/A**

Table 3. *Energy ranges and filter configurations of the 'Niton XL3t Ultra' XRF analyser when operated in "Mining" and "Soil" modes.*

Sample	**Ti**	**Ti ±**	**Fe**	**Fe ±**	**Zn**	**Zn ±**	**Rb**	**Rb ±**	**Nb**	**Nb ±**	**Sr**	**Sr ±**	**Y**	**Y ±**	**Zr**	**Zr ±**	***Th***	***Th ±***	***U***	***U ±***	**Source**
UC.01	486	10.8	7542	40.8	25	2.4	225	1.4	12	0.9	61	0.7	61	1.6	81	1.9	*n.r.*		*n.r*		**C1**
UC.02	489	10.4	7684	40.2	23	2.4	209	1.4	10	0.9	69	0.7	56	1.4	82	1.9	*n.r.*		*n.r*		**C1**
UC.03	553	9.6	6847	41.6	25	2.6	181	1.4	8	0.9	60	0.7	42	1.5	76	2.0	*n.r.*		*n.r*		**C1**
TL.01	719	12.4	10568	48.6	43	2.7	203	1.4	13	0.9	75	0.7	50	1.4	149	2.2	*23*	*0.9*	*1*	*1.3*	**C2T**
TL.02	1138	13.8	12660	56.3	40	2.8	212	1.5	12	1.0	84	0.8	51	1.5	186	2.5	*24*	*0.9*	*6*	*1.5*	**C2E**
TL.03	1112	14.0	12875	57.9	41	2.9	210	1.6	10	1.0	81	0.8	47	1.5	177	2.5	*22*	*0.9*	*7*	*1.5*	**C2E**
TL.04	1102	13.6	12292	57.1	38	2.9	212	1.6	10	1.0	84	0.9	47	1.5	185	2.6	*26*	*0.9*	*8*	*1.5*	**C2E**
TL.05	709	12.1	11442	52.3	44	2.8	203	1.5	13	0.9	82	0.8	50	1.4	142	2.3	*23*	*0.9*	*6*	*1.3*	**C2T**
TL.06	1135	13.8	12846	55.3	35	2.7	215	1.5	13	0.9	83	0.8	52	1.5	183	2.4	*20*	*0.9*	*12*	*1.4*	**C2E**
TL.07	1057	13.5	11567	53.4	30	2.7	196	1.5	9	0.9	76	0.8	48	1.6	170	2.3	*24*	*0.9*	*15*	*1.4*	**C2E**
TL.08	1144	14.7	12984	56.3	33	2.7	217	1.5	13	1.0	83	0.8	56	1.6	186	2.4	*21*	*0.8*	*11*	*1.4*	**C2E**
TL.09	1129	14.1	12992	55.2	38	2.7	218	1.5	12	0.9	84	0.8	54	1.5	186	2.4	*22*	*0.9*	*13*	*1.4*	**C2E**
TL.10	1144	14.2	13361	56.5	43	2.8	216	1.5	11	0.9	86	0.8	51	1.5	187	2.4	*22*	*0.9*	*11*	*1.5*	**C2E**
TL.11	1323	17.1	14983	58.5	39	2.7	249	1.6	16	1.0	99	0.9	67	1.7	208	2.4	*25*	*0.9*	*11*	*1.5*	**C2E**
TL.12	1324	16.9	15530	58.8	41	2.6	248	1.5	16	0.9	100	0.8	66	1.6	205	2.4	*23*	*0.9*	*10*	*1.5*	**C2E**
TL.13	1371	17.5	15336	59.7	35	2.6	250	1.6	17	1.0	100	0.9	66	1.6	207	2.5	*23*	*0.9*	*10*	*1.5*	**C2E**
TL.14	1240	16.0	14126	56.1	40	2.6	235	1.5	15	0.9	92	0.8	63	1.6	200	2.4	*25*	*0.9*	*12*	*1.5*	**C2E**
TL.15	1162	14.2	12997	56.6	41	2.8	216	1.5	11	1.0	87	0.8	48	1.4	187	2.4	*24*	*0.9*	*16*	*1.5*	**C2E**
TL.16	1201	14.8	13335	54.1	38	2.6	222	1.5	11	0.9	90	0.8	56	1.5	193	2.3	*23*	*0.8*	*11*	*1.4*	**C2E**
TL.17	1133	14.4	12809	53.6	35	2.6	215	1.4	12	0.9	87	0.8	55	1.5	189	2.3	*23*	*0.9*	*14*	*1.4*	**C2E**

Sample	**Ti**	**Ti ±**	**Fe**	**Fe ±**	**Zn**	**Zn ±**	**Rb**	**Rb ±**	**Nb**	**Nb ±**	**Sr**	**Sr ±**	**Y**	**Y ±**	**Zr**	**Zr ±**	***Th***	***Th ±***	***U***	***U ±***	**Source**
TL.18	1132	14.3	12785	53.9	38	2.6	216	1.5	12	0.9	84	0.8	55	1.5	188	2.3	*22*	*0.8*	*14*	*1.4*	**C2E**
TL.19	1202	15.1	13563	54.7	38	2.6	226	1.5	13	0.9	90	0.8	59	1.5	194	2.3	*24*	*0.9*	*17*	*1.5*	**C2E**
TL.20	1175	14.7	12941	53.5	32	2.5	216	1.4	13	0.9	86	0.8	57	1.5	189	2.3	*25*	*0.9*	*14*	*1.4*	**C2E**
TL.21	1181	14.9	12348	53.5	34	2.6	211	1.5	11	0.9	85	0.8	54	1.5	183	2.3	*21*	*0.8*	*17*	*1.4*	**C2E**
TL.22	1205	14.4	13047	54.5	35	2.6	216	1.5	11	0.9	86	0.8	53	1.5	191	2.4	*n.r.*		*n.r.*		**C2E**
TL.23	516	10.9	9025	43.9	33	2.5	234	1.5	12	0.9	72	0.7	61	1.5	96	2.0	*16*	*0.8*	*15*	*1.4*	**C1**
TL.24	537	11.7	8233	43.6	25	2.5	227	1.5	13	0.9	70	0.7	63	1.6	94	2.0	*12*	*0.8*	*18*	*1.4*	**C1**
TL.25	650	11.5	7673	41.3	29	2.5	186	1.3	6	0.9	79	0.8	48	1.4	85	1.9	*16*	*0.8*	*17*	*1.4*	**C1**
TL.26	483	9.7	7365	42.1	26	2.5	203	1.4	7	0.9	65	0.7	45	1.4	79	2.0	*15*	*0.8*	*21*	*1.4*	**C1**
TL.27	1076	13.6	11702	53.9	38	2.8	195	1.5	10	0.9	78	0.8	48	1.6	181	2.4	*23*	*0.9*	*11*	*1.4*	**C2E**
TL.28	1159	14.9	12857	55.1	40	2.7	217	1.5	12	0.9	88	0.8	52	1.4	193	2.4	*23*	*0.9*	*10*	*1.4*	**C2E**
TL.29	1141	15.7	12994	54.3	38	2.6	218	1.5	12	0.9	90	0.8	59	1.5	193	2.3	*25*	*0.9*	*7*	*1.4*	**C2E**
TL.30	1166	14.2	12976	54.9	36	2.6	220	1.5	10	0.9	86	0.8	51	1.4	190	2.4	*22*	*0.8*	*8*	*1.4*	**C2E**
TL.31	1119	14.2	12451	56.9	40	2.9	205	1.5	9	1.0	82	0.8	49	1.5	179	2.4	*24*	*0.9*	*9*	*1.4*	**C2E**
TL.32	1331	16.9	16149	61.3	40	2.7	248	1.6	16	1.0	97	0.9	69	1.7	214	2.5	*25*	*0.9*	*6*	*1.4*	**C2E**
TL.33	1177	14.6	12926	55.5	38	2.7	212	1.5	11	0.9	87	0.8	55	1.5	197	2.4	*23*	*0.9*	*10*	*1.4*	**C2E**
TL.34	733	13.4	10285	48.6	43	2.7	195	1.4	12	0.9	80	0.8	48	1.4	144	2.2	*22*	*0.8*	*8*	*1.3*	**C2T**
TL.35	1234	15.5	13984	56.0	38	2.7	229	1.5	13	0.9	92	0.8	61	1.5	194	2.4	*22*	*0.8*	*11*	*1.4*	**C2E**
TL.36	1192	15.2	13085	54.8	32	2.6	216	1.5	11	0.9	87	0.8	57	1.6	187	2.3	*24*	*0.9*	*10*	*1.4*	**C2E**
TL.37	756	12.2	10248	49.4	44	2.8	197	1.4	11	0.9	76	0.8	48	1.4	138	2.2	*23*	*0.8*	*3*	*1.3*	**C2T**
TL.38	1168	14.6	12899	54.8	40	2.7	219	1.5	12	0.9	87	0.8	52	1.4	193	2.4	*21*	*0.8*	*8*	*1.4*	**C2E**

Sample	Ti	Ti ±	Fe	Fe ±	Zn	Zn ±	Rb	Rb ±	Nb	Nb ±	Sr	Sr ±	Y	Y ±	Zr	Zr ±	*Th*	*Th ±*	*U*	*U ±*	Source
TL.39	1173	15.5	12772	54.7	30	2.6	230	1.5	14	1.0	87	0.8	63	1.6	196	2.4	*25*	*0.9*	*7*	*1.5*	**C2E**
TL.40	1439	18.7	16567	64.8	47	2.9	264	1.7	19	1.0	102	0.9	74	1.8	215	2.6	*26*	*0.9*	*7*	*1.5*	**C2E**
TL.41	1305	16.4	14177	57.5	33	2.6	233	1.5	13	0.9	93	0.8	61	1.6	196	2.4	*23*	*0.9*	*6*	*1.5*	**C2E**
TL.42	1353	16.0	14048	56.3	37	2.6	225	1.5	12	0.9	89	0.8	59	1.5	193	2.4	*21*	*0.8*	*9*	*1.4*	**C2E**
TL.43	980	12.1	10291	52.2	37	2.9	187	1.5	9	1.0	78	0.8	43	1.5	175	2.4	*25*	*0.9*	*11*	*1.5*	**C2E**
TL.44	1303	16.2	14345	57.4	34	2.6	232	1.5	15	0.9	94	0.8	64	1.6	200	2.4	*23*	*0.9*	*10*	*1.4*	**C2E**
TL.45	1560	20.7	18389	79.3	41	3.4	269	2.0	20	1.2	107	1.1	71	2.2	215	3.0	*27*	*1.2*	*5*	*1.9*	**C2E**
TL.46	1523	19.7	18659	71.8	47	3.2	270	1.8	20	1.1	105	1.0	70	1.9	215	2.8	*26*	*1.0*	*10*	*1.7*	**C2E**
TL.47	1224	15.5	13848	55.2	34	2.5	228	1.5	13	0.9	92	0.8	61	1.5	200	2.4	*22*	*0.9*	*10*	*1.4*	**C2E**
TL.48	1229	15.1	13644	56.5	38	2.6	223	1.5	12	0.9	89	0.8	55	1.4	194	2.4	*23*	*0.9*	*12*	*1.4*	**C2E**
TL.49	1215	14.9	13405	54.5	34	2.6	223	1.5	12	0.9	92	0.8	56	1.5	191	2.3	*20*	*0.8*	*0*	*1.4*	**C2E**
TL.50	1180	16.1	12884	56.3	35	2.7	216	1.5	14	1.0	86	0.8	61	1.7	190	2.4	*23*	*0.9*	*0*	*1.4*	**C2E**
TL.51	1206	14.6	12610	54.5	40	2.7	206	1.5	10	0.9	87	0.8	51	1.5	191	2.4	*23*	*0.9*	*6*	*1.4*	**C2E**
TL.52	1376	18.3	15710	64.6	44	3.0	247	1.7	17	1.0	100	0.9	69	1.9	208	2.6	*28*	*1.0*	*3*	*1.6*	**C2E**
TL.53	1213	15.3	13200	54.4	34	2.6	217	1.5	13	0.9	87	0.8	57	1.5	189	2.3	*21*	*0.8*	*0*	*1.4*	**C2E**
TL.54	1516	19.4	17123	66.3	49	2.9	265	1.7	19	1.0	103	0.9	72	1.8	215	2.6	*24*	*0.9*	*4*	*1.5*	**C2E**
TL.55	1330	16.7	14552	56.4	45	2.7	237	1.5	13	0.9	95	0.8	64	1.6	205	2.4	*24*	*0.9*	*5*	*1.5*	**C2E**
TL.56	1165	14.9	12378	55.3	35	2.7	207	1.5	11	1.0	83	0.8	53	1.6	185	2.4	*22*	*0.9*	*3*	*1.4*	**C2E**
TL.57	1218	15.3	13048	55.2	38	2.7	217	1.5	12	0.9	86	0.8	58	1.6	191	2.4	*23*	*0.8*	*3*	*1.4*	**C2E**

Table 4. *Element concentrations measured in obsidian artefacts from Uliești – Croitori (UC) and Măgura – Buduiasca (TL) by XRF, after adjustment using calibration factors derived from corresponding measurements on certified reference materials (CRMs). The XRF analyser was operated in "Soil mode" for measuring Th and U, and in "Mining mode" for all other elements. Values (μ ± 1σ) are given in parts per million (ppm). Abbreviations: C1 – Carpathian 1; C2E – Carpathian 2E; C2T – Carpathian 2T; n.r. – not recorded.*

DISCUSSION

Our analysis of the material from Uliești – Croitori and Măgura – *Buduiasca* is not the first geochemical provenancing study of the obsidian assemblages from these sites, but it goes beyond previous research and leads to rather different conclusions.

pXRF analysis of the four obsidian artefacts from Uliești and Corbii Mari by the late Dr Bogdan Constantinescu of the "Horia Hulubei" National Institute for Research and Development in Physics and Nuclear Engineering (IFIN-HH) in Bucharest identified the geological provenance of the obsidian as the Carpathian 1 source area near Viničky in southeast Slovakia (reported in Ilie, Niță 2014). Constantinescu used an Oxford Instruments 'X-MET 3000-TX' handheld analyser to obtain XRF raw data in the form of a spectrum graph (Ilie, Niță 2014, pl. 4), which identifies which elements are present in the sample but not how much of each element is present. In this sense, the raw spectrum is "qualitative" data, and visual assessment of a spectrum may not be sufficient to establish the exact provenance of a sample.

The Niton 'XL3t Ultra' analyser used in our research is a more modern instrument, in which raw spectrum data are processed mathematically in software to yield concentration values for the various elements detected in a sample, and such 'quantitative' data can be further refined by calibration using external reference standards. Our results (Table 4; Fig. 5) indicate that the three obsidian artefacts from Uliești – Croitori do indeed originate from a geological source in the Western Carpathians, and almost certainly from the Carpathian 1 (C1) source area in southeast Slovakia. One sample falls outside the C1 source ellipse in Fig. 5; while this may be an aberrant measurement, it is more likely that the ellipse underestimates the range of variation in C1 obsidian being based on elemental data for just 15 geological reference samples.

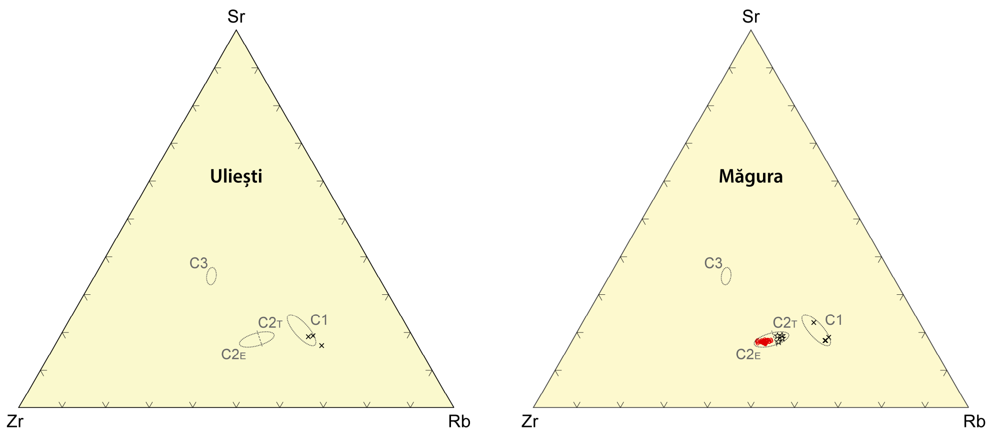

Figure 5. *Ternary graph of Zr/Sr/Rb data (Table 4) for obsidian artefacts from Uliești – Croitori and Măgura – Buduiasca, plotted against the compositional ranges (ellipses) of obsidian reference samples from sources in the Carpathians.*

Kasztovsky *et alii* (2019) analysed five obsidian artefacts from Măgura – *Buduiasca*, which they classified as core fragments (*n* = 3) and flakes (*n* = 2). These were attributed to the earliest (Starčevo-Criș I) occupation phase, but no additional information on their context or find locations was provided. Based on the results of Prompt Gamma-Ray Activation Analysis (PGAA) the geological source area of the obsidian was determined as northeast Hungary (Carpathian 2). Based on their Titanium (Ti) content, two samples could be assigned to the C2T (Tolcsva vicinity) source, and two to the C2E (Mád-Erdőbénye) source.

Our analyses provide further detail on the use of obsidian at Măgura – *Buduiasca*. Of 57 obsidian pieces analysed, 53 were excavated from the *Boldul lui Moș Ivănuș* area of the site, and all are likely to date to the earliest (Starčevo-Criș I) occupation phase based on archaeostratigraphic and radiocarbon evidence – at least none of the obsidian from this area can be shown to date to the later Early Neolithic (Starčevo-Criș III phase). The XRF data show that the great majority (*n* = 49) of these finds originated from the C2E source with the remainder (*n* = 4) deriving from the C2T source. The five pieces from Măgura – *Buduiasca* analysed by Kasztovsky *et alii* (2019) correspond to samples TL.01–TL.05 in our series (Table 4). Our XRF data for these samples are entirely consistent with those obtained using PGAA although, whereas we were able to assign all five samples to source (C2E or C2T) based on the Sr, Zr, Rb (confirmed by the Ti and Fe) values, Kasztovsky *et alii* (2019) were only able to discriminate between C2E and C2T in four cases. The four obsidian specimens included in our analysis that were not found in the *Boldul lui Moș Ivănuș* area probably all relate to the Starčevo-Criș III occupation phase and proved to be from a C1 (southeast Slovakia) source. On this evidence, it would seem there was a shift in obsidian use at Măgura – *Buduiasca* from C2 obsidian in the early part of the Starčevo-Criș period between 6000–5800 cal BC to C1 obsidian in the later part of the Early Neolithic between 5800–5600 cal BC.

The question arises whether this temporal trend was a wider regional phenomenon? As noted in the *Introduction*, obsidian has been found at only a small number of Early Neolithic sites in the Lower Danube basin (Fig. 1) and in most cases only in very small quantities, while geochemical fingerprinting has been attempted at only five of these sites – Uliești, Corbii Mari and Măgura – *Buduiasca* in Romania (this paper; Ilie, Niță 2014) and Ohoden – *Valoga* and Dzhulyunitsa – *Smardesh* in Bulgaria (Bonsall *et alii* 2017). C2 obsidian occurs at three of the sites, but the chronological context arguably is only securely established at Măgura – *Buduiasca* and Dzhulyunitsa, where large series of AMS ^{14}C dates place the C2 obsidian finds in the period before 5800 cal BC. At Ohoden the two finds of C2E obsidian were 'attributed' to a period after 5800 cal BC based on the excavator's interpretation of the site stratigraphy and ceramic typology (Bonsall *et alii* 2017), but no details of the find locations or archaeological contexts are available for those finds and there are no associated radiocarbon dates. C1 obsidian has also been documented at four of the sites in our study region, although the chronology is no less ambiguous. At Măgura – *Buduiasca* finds of C1 obsidian seem securely dated to the late Early Neolithic. There are no ^{14}C dates for Uliești and Corbii Mari where "dating" of the C1 obsidian rests on its co-occurrence with Starčevo-Criș III pottery in a low-density surface scatter. Likewise, at Ohoden in northern Bulgaria dating of the C1 obsidian to the late Starčevo period rests on ceramic typology. Taken together, this evidence is at least consistent with the results of obsidian provenance studies in other regions of Romania that point to preferential use of C1 obsidian throughout Romania from the later stages of the Early Neolithic onwards (Glascock *et alii* 2017; Boroneanț *et alii* 2018).

Our XRF measurements on the Măgura – *Buduiasca* material were undertaken in the course of two one-day visits to the Teleorman County Museum in Alexandria by AB and CB. Time did not permit more than basic recording of the individual pieces, which involved photographing each piece, measuring their dimensions and weights, and low-level typological classification. No use-wear analysis or refitting could be undertaken. Nevertheless, the information obtained yields some clues to the treatment of obsidian by the Early Neolithic inhabitants of the site. A striking feature is the disparity in the number of pieces recovered from *Boldul lui Moș Ivănuș* compared to the rest of the site. Yet the total area excavated at *Boldul lui Moș Ivănuș* was smaller (168 m^2 compared to 238 m^2) and methods of excavation and recovery (which included routine dry sieving using a standard 10 mm mesh size) were essentially the same in all parts of the site. The obsidian frequency data therefore imply greater use or availability of obsidian during the Starčevo-Criș I occupation phase and a sharp reduction in obsidian use in the ensuing Starčevo-Criș III phase, coincident with a change in the procurement pattern from C2 to C1 obsidian. From a techno-typological perspective, blades/bladelets (or fragments thereof) make up a surprisingly high percentage (ca. 34%) of the C2 obsidian artefacts from *Boldul lui Moș Ivănuș*. Yet, there are no residual blade cores and very few primary (corticated) removals in the assemblage. If this is a representative sample, then these characteristics suggest that C2 obsidian may have reached the site mainly in the form of blade and flake blanks (Fig. 3/1–3), rather than as raw nodules (*n.b.* the three "core fragments" mentioned by Kasztovsky *et alii* 2019 were classified by us as 'irregular flakes').

CONCLUSIONS

The research presented in this paper forms part of a comprehensive study of obsidian distribution patterns in Romanian prehistory with the overall aim of establishing the patterns of movement, modes of acquisition and use of obsidian during different archaeological periods. Though based on assemblages from just two sites in the province of Muntenia, this nevertheless represents the most detailed obsidian provenancing study to date relating to the Early Neolithic in southern Romania.

All the obsidian analysed originated from sources in the Western Carpathians, and predominantly from the Carpathian 2 source area in northeast Hungary. The vast majority of the obsidian from the earliest Neolithic (Starčevo-Criș I) occupation of the Măgura – *Buduiasca* site derives from the C2E source, while a small proportion of the obsidian from the same occupation phase at Măgura came from the C2T source.

The importance of the Măgura – *Buduiasca* site for obsidian provenance studies lies in its long and detailed Neolithic sequence, comprising four main occupation phases: early Starčevo, late Starčevo, Dudești and Vădastra. Most of the obsidian artefacts from the site relate to the earliest phase and the raw material came exclusively from C2 sources. A change is evident in the late Early Neolithic with an apparent decline in obsidian use and a shift toward acquisition of material ultimately from the C1 source area in southeast Slovakia. Though less satisfactory, the evidence from Uliești also points to late Early Neolithic use of C1 obsidian. Interestingly, no obsidian was recovered from Middle (Dudești) or Late (Vădastra) Neolithic contexts at Măgura.

Comparison of our elemental composition results with those from a previous study of obsidian from Măgura – *Buduiasca* shows that pXRF offers a very effective alternative to non-destructive, but much more expensive, laboratory-based techniques like PGAA for obsidian provenance studies. It also highlights the importance of analysing large, representative series of obsidian artefacts in order to adequately characterize the range of obsidian types present in an assemblage – a task that can be accomplished very easily with pXRF.

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